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(54) Title: REMOVAL OF DRAG REDUCER ADDITIVE FROM FUEL BY TREATMENT WITH SELECTED ACTIVATED CARBONS AND GRAPHITES

(57) Abstract: The application relates to a method for selecting drag reducer additive (DRA) effectively removable by activated carbons and graphites to be used in fuel. The application also relates to effective activated carbons and graphites for removing DRA from fuel, and to a method of using effective activated carbons and graphites to remove DRA from fuel.

**TITLE: REMOVAL OF DRAG REDUCER ADDITIVE FROM FUEL BY
TREATMENT WITH SELECTED ACTIVATED CARBONS
AND GRAPHITES**

5 **Cross-reference to Related Applications**

[0001] This application is a continuation-in-part application of U.S. Patent Application No. 10/124,974, filed on April 18, 2002.

Field of the Invention

[0002] The application relates to a method for selecting drag reducer additive
10 (DRA) effectively removable by activated carbon to be used in fuel. The application also relates effective activated carbons and graphites for removing DRA from fuel, and to a method of using effective activated carbon and graphites to remove DRA from fuel.

Background

15 [0003] In order to move fluid through pipelines, into or out of wells, or through equipment, energy must be applied to the fluid. The energy moves the fluid, but is lost in the form of friction. This frictional pressure drop, or drag, restricts the fluid flow, limiting throughput and requiring greater amounts of energy for pumping.

[0004] Materials can be added to flowing fluids in order to reduce the energy lost
20 due to friction, or drag, thus permitting the movement of more fluid at the same differential pressure. The resulting reduction in frictional pressure drop improves pumping efficiency, lowers energy costs, and increases profitability. Materials for reducing drag in flowing fluids are generally known by the generic names "flow improver" or "drag reducer additive" (sometimes referred to as "DRA").

[0005] Unfortunately, whether in the virgin form or in the sheared or partially sheared form, and despite the fact that it is intentionally added to certain fuels, drag reducer additive nonetheless is a "contaminant" in liquid hydrocarbon fuels, and has the potential to cause a number of problems. For example, the presence of drag
5 reducer additive in motor gasoline, even in the sheared form, has caused increased intake valve deposits, plugging of fuel filters, and increased combustion chamber deposits. In diesel fuels, drag reducer additive may cause plugging of fuel filters and strainers and increased fuel injector deposits. Drag reducer additive is prohibited in aviation turbine fuels, although it has been observed as a contaminant due to
10 accidental addition or other non-intentional means. The presence of drag reducer additive in aviation turbine fuel may result in downgrading of the entire batch to non-aviation kerosene or diesel fuel, both of which generally have less market value.

[0006] Viable methods of detecting and quantifying drag reducer additive in liquid hydrocarbon fuels commonly employ gel permeation chromatography, which is time
15 consuming and expensive. Because of this, contaminated liquid hydrocarbon fuels often are used, despite the potential problems if drag reducer additive is present. Contaminated aviation turbine fuels may be diverted to other uses or returned to a refinery for reprocessing, either of which results in additional expense. Simple and inexpensive methods and materials are needed for removing drag reducer additive
20 from liquid hydrocarbon fuels.

Summary

[0007] The application provides a method for removing drag reducer additive from

a liquid hydrocarbon fuel. The method comprises providing a contaminated liquid hydrocarbon fuel comprising a concentration of a drag reducer additive, and contacting the contaminated liquid hydrocarbon fuel with a quantity of one or more of an activated carbon or a graphite effective to substantially reduce said concentration of drag reducer additive, thereby producing a clean liquid hydrocarbon fuel.

Brief Description of the Figures

- [0008] Figure 1 is a graph of % Polymer Removed vs. Carbon (activated carbons and graphites) used to remove unsheared FLO[®]XS polymer from jet fuel using the Grad Add/Stir method (described in detail below).
- 10 [0009] Figure 2 is a graph of % Polymer Removed vs. Carbon (activated carbons and graphites) used to remove unsheared FLO[®]XS polymer from jet fuel for both Grad Add/Stir and Quick Add/Stir methods using the Grad Add/Stir method.
- [0010] Figure 3 is a graph of % Polymer Removed vs. Carbon (activated carbons and graphites) used to remove sheared FLO[®]XS polymer from jet fuel using the Grad
- 15 Add/Stir method.
- [0011] Figure 4 is a graph of % Polymer Removed vs. Carbon (activated carbons and graphites) used to remove sheared FLO[®]XS polymer from jet fuel for both Grad Add/Stir and Quick Add/Stir methods.
- [0012] Figure 5 is a graph of % Polymer Removed vs. Carbon (activated carbons
- 20 and graphites) used to remove FLO[®]XS polymer from jet fuel for Unsheared vs. Sheared FLO[®]XS polymer using the Grad Add/Stir method.

Detailed Description

[0013] The application provides methods for removing drag reducer additive (DRA) from liquid hydrocarbon fuels, preferably motor gasoline or jet fuel, most preferably jet fuel, using selected activated carbons or graphites. As used herein, the word “contaminated” refers to the presence of DRA in the fuel, due to either intentional addition or unintentional addition.

“Liquid Hydrocarbon Fuel”

[0014] By “liquid hydrocarbon fuel” is meant any hydrocarbon that is liquid under conditions of transport and/or storage. Suitable liquid hydrocarbon fuels include, but are not necessarily limited to those having a boiling range of from about 150 °F to about 750 °F, which may be used as a fuel. In one embodiment, the liquid hydrocarbon fuel is selected from the group consisting of liquefied natural gas (LNG), liquefied petroleum gas (LPG), motor gasoline, aviation gasoline, distillate fuels such as diesel fuel and home heating oil, kerosene, jet fuel, No. 2 oil, residual fuel, No. 6 fuel, or bunker fuel. In a preferred embodiment, the liquid hydrocarbon fuel is selected from the group consisting of diesel fuel, jet fuel, aviation gasoline, and motor gasoline. In a more preferred embodiment, the liquid hydrocarbon fuel is jet fuel, at least in part due to the stringent requirements applicable to jet fuel and drag reducer additive. The phrase “jet fuel” refers to both commercial jet fuel (Jet A, Jet A-1, and JET B) and military jet fuel, such as JP-4, JP-5, JP-8 and the like.

“Drag Reducer Additive”

[0015] The term “drag reducer additive” (i.e. DRA) is defined to mean polyolefin

polymers comprising polyolefin moieties which are introduced into petroleum liquids for the purpose of reducing fluid flow drag. The drag reducer additive may comprise other components besides the polyolefin moieties. Examples of such components include, but are not necessarily limited to surfactant, catalyst residue, other additives, and other byproducts from the production of the polymer. The polymer itself may contain other non-olefin monomer units as well.

[0016] In a preferred embodiment, the drag reducer additive includes, but is not necessarily limited to, non-polar long-chain polyolefin polymers, generally referred to as "polyalphaolefins," having a "peak" molecular weight sufficiently high to allow the polymers to reduce fluid flow drag. Suitable polyalphaolefins are believed to have a molecular weight of about 1 million Daltons or more, more preferably about 10 million Daltons or more, most preferably about 25 million Daltons or more. The "peak" molecular weight refers to the peak that typically is measured as the drag reducer is eluted and detected during gel permeation chromatography.

[0017] Suitable polyalphaolefins comprise polymerized linear alpha olefin (LAO) monomers having from about 2 to about 40 carbon atoms, preferably from about 2 to about 30 carbon atoms, more preferably from about 4 to about 20 carbon atoms, most preferably from about 6 to about 12 carbon atoms. An especially preferred embodiment for a DRA which is effectively removable by the activated carbons and/or graphites described herein comprises at least two different LAO's, preferably having from about 6 to about 12 carbon atoms, the number of carbon atoms of the "at least two different LAO's" differing by 6.

[0018] Polyalphaolefins having relatively high molecular weights are required to impart good drag reduction. Suitable polyalpha olefins "are made by a variety of processes, including but not necessarily limited to solution polymerization and bulk polymerization. Bulk polymerization is said to produce "ultra-high molecular weight polyolefin drag reducers [that] are significantly larger (molecular weight basis) than the best molecular weights made by solution polymerization." See U.S. Patent No. 5,504,132. Preferred DRA's for removal according to the process described herein are made by solution polymerization.

[0019] Without limiting the invention to a specific theory or mechanism of action, the very large polyalpha olefins made by bulk polymerization may be more difficult to adsorb onto and retain on the carbonaceous removal agents. In contrast, the polyalpha olefins made by solution polymerization may be more readily adsorbable onto the removal agents, and more readily retained by the removal agents.

[0020] Drag reducer additives are generally unsheared, partially sheared, or fully sheared. An additive that is fully sheared is one that is degraded in molecular weight to the maximum extent possible using high shear devices such as pumps, static mixers, etc. Commercially available drag reducer additives include, but are not necessarily limited to, CDR[®] Flow Improver and REFINED POWER[™], manufactured by Conoco Specialty Products, Inc., EN-660 Flow Improver, manufactured by Energy 2000 LLC, and FLO[®]XS and FLO[®]XL, manufactured by Baker Petrolite. In a preferred embodiment, the drag reducer additive is FLO[®]XS and equivalents thereof.

[0021] The exact mechanism by which a drag reducer additive reduces drag in flowing liquid hydrocarbons is not completely known. However, a drag reducer additive apparently alters the turbulent flow regime of the liquid hydrocarbons. In a pipeline, this flow regime is comprised of at least three regions. At the center of the pipe is a turbulent core, which is the largest region and includes most of the fluid in the pipe. This is the zone of eddy currents and random motions for which turbulent flow is named. Nearest to the pipe line wall is the laminar sublayer. In this zone, the fluid moves laterally in "sheets". Between the laminar layer and the turbulent core lies the "buffer zone". It appears that much of the turbulence which exists in turbulent flow develops when a portion of the laminar sublayer, called a "streak", moves up to the buffer zone, where it begins to vortex and oscillate, finally breaking up and throwing fluid into the core. This ejection of fluid into the core is called a "burst". The burst creates the turbulence in the core, and energy is wasted in different directions. Drag reducer additive appears to interfere with the bursting process and prevent or reduce the degree of turbulence by stretching in the flow, absorbing the energy in the streak, and thereby preventing bursts.

[0022] As liquids containing drag reducer additive travel through pumps, pipelines and other equipment, the drag reducer additive typically degrades through shearing action, resulting in a reduction in the molecular weight of the drag reducer additive. The degraded drag reducer additive is generally sheared or partially sheared drag reducer additive. Upon reaching the ultimate destination, liquid hydrocarbon fuels that have been shipped using drag reducer additive may contain a significant amount

of drag reducer additive, including that in the sheared and partially sheared form.

Removal Agents for Removing DRA from Fuels

[0023] The present application is based on the surprising finding that selected activated carbons and graphites, particularly selected graphites, are much more effective than others as removal agents for binding and removing drag reducer additive from fuels, preferably motor gasoline and jet fuels, most preferably jet fuels. The superiority has been demonstrated in unsheared DRA known as Baker Petrolite FLO[®]XS.

[0024] Without limiting the application to a particular theory or mechanism of operation, the more active removal agents are believed to comprise ducts or pores having a hydrophobic/hydrophilic property that provides a chemical attraction to pendant groups on the drag reducer additive. The chemical attraction is believed to bring the pendant groups on the drag reducer additive into proximity and orientation with the pore surface of the agent, thereby immobilizing the pendant groups. Because many pendant groups on a given drag reducer molecule are simultaneously immobilized, the drag reducer molecule is strongly immobilized.

[0025] Without limiting the claims to a particular mechanism or theory of action, the effective activated carbons and graphites are believed to be porous materials comprising pores having a hydrophobic/hydrophilic property that is compatible with or provides a chemical attraction to pendant groups of the particular drag reducer additive. The specific activated carbons and graphites may be in the form of crushed particles or granules, powder, cylinders, globules, fibers, or honeycombs. Preferred

agents are in the form of particles or granules. Most preferred agents are in powder or granule form.

Activated Carbons

[0026] Suitable activated carbons are commercially available, for example, from

5 Allchem Industries, Inc., Beta Chemicals, Calgon, Coyne Chemical Co., Elf Atochem North America, Inc. (Performance Products), R. W. Greef & Co, Inc., Kingshine Chemical Co., Ltd., Mays Chemical Co., Inc., Mitsubishi International Corp. (Industrial Specialty Chemicals Div.), Spectrum Chemical Mfg. Corp., Norit and others. When added (in increments with agitation) to a fuel mixture containing a

10 preferred unsheared drag reducer additive of about 8-12 ppm polymer concentration, more preferably about 9-11 ppm polymer concentration, most preferably about 10 ppm polymer concentration, suitable activated carbons attain a % polymer removal of about 20% or more; preferably about 30% or more; more preferably at least about 40% or more, at about 1g activated carbon/100 ml fuel. This equates to a %

15 adsorption capacity of about 0.014% or more, preferably about 0.02% or more, most preferably about 0.03% or more. When added (in increments with agitation) to a fuel mixture containing a preferred sheared drag reducer additive of about 8-12 ppm polymer concentration, more preferably about 9-11 ppm polymer concentration, most preferably about 10 ppm polymer concentration, suitable activated carbons attain a %

20 polymer removal of about 20% or more; preferably about 25% or more; more preferably about 30% or more, at about 1g activated carbon/100 ml fuel. This equates to a % adsorption capacity of about 0.018% or more, preferably about 0.025% or

more, more preferably about 0.03% or more.

[0027] Commercially viable activated carbons, which have been demonstrated to be suitable to remove Baker Petrolite FLO[®] XS and equivalents thereof include, but are not necessarily limited to, CALGON ADP, CALGON COLORSORB, CALGON WPX, NORIT A SUPRA, NORIT CA 1, NORIT FGD, NORIT HDB, SXO POWDER, and CARBON 5565. Preferred activated carbons demonstrated to be useful for removing Baker Petrolite FLO[®] XS and equivalents thereof include, but are not necessarily limited to CALGON WPX, NORIT A SUPRA, NORIT CA1, NORIT FGD, NORIT HDB, SXO POWDER and CARBON 5565. Most preferred activated carbons demonstrated to be useful for removing Baker Petrolite FLO[®] XS and equivalents thereof include, but are not necessarily limited to NORIT A SUPRA, NORIT CA1, NORIT FGD, and NORIT HDB.

Preferred Graphites

[0028] Most preferred carbonaceous materials are graphites. Graphite is a crystalline form of carbon found as a naturally occurring mineral in many locations around the world. Graphite can be amorphous ("amorphous graphite"). Graphite also can have a perfect basal cleavage which, coupled with its extreme softness, gives it an oily, slippery feel. such graphites include, but are not necessarily limited to natural graphite, synthetic graphite, and expanded graphite. Each of these graphite types is commercially available in various forms, including, crystalline lumps, crystalline large flakes, crystalline medium flakes, crystalline small flakes, and powder form. Artificial graphite can be manufactured from petroleum coke and is

primarily used to make electrodes. The virgin by-product of such electrode production has a carbon content as high as 99.9%, and can be a relatively inexpensive source of graphite agent, to highly refined natural graphite. Suitable candidate graphites are commercially available, for example, from Asbury Carbons, Inc.,
5 Asbury, NJ; Superior Graphite Co., Chicago, IL; Stanford Materials Corporation, Aliso Viejo, CA; and others.

[0029] Preferred graphites comprise graphite powders or granular graphite particulates. The granular graphite particulates have an average diameter of from about 0.01 microns to about 10,000 microns; preferably from about 0.1 microns to
10 about 1,000 microns; most preferably about 1 micron to about 100 microns. Preferred graphites have a porosity sufficient to provide an adsorption capacity of about 0.01 wt.% or more, preferably about 0.03 wt.% or more, most preferably about 0.04 wt%, when added to a preferred drag reducer additive. Suitable and preferred graphites are commercially available from Superior Graphite Company. Preferred graphite
15 products comprise, but are not necessarily limited to, purified carbon, natural graphite, silica (crystalline quartz), and synthetic graphite.

[0030] When added (in increments with agitation) to a fuel mixture containing a preferred unsheared drag reducer additive of about 8-12 ppm polymer concentration, more preferably about 9-11 ppm polymer concentration, most preferably about 10
20 ppm polymer concentration, suitable graphites attain a % polymer removal of about 30% or more; preferably about 40% or more; more preferably at least about 50% or more, at about 1g activated carbon/100 ml fuel. This equates to a % adsorption

capacity of about 0.02% or more, more preferably about 0.03% or more, most preferably about 0.04% or more. When added (in increments with agitation) to a fuel mixture containing a preferred sheared drag reducer additive of about 8-12 ppm polymer concentration, more preferably about 9-11 ppm polymer concentration, most preferably about 10 ppm polymer concentration, suitable graphites attain a % polymer removal of at least about 25%; more suitably at least about 30%; most suitably at least about 35%, at about 1g activated carbon/100 ml fuel. This equates to a % adsorption capacity of about 0.02%, more preferably about 0.025%, most preferably about 0.03%.

10 [0031] Graphites that demonstrated commercial viability for adsorbing unsheared and sheared BAKER PETROLITE FLO[®] XS and equivalents included GRAPHITE 2126, GRAPHITE 2139, GRAPHITE 3726, GRAPHITE 3739, GRAPHITE 5526, GRAPHITE 5539, GRAPHITE 9026, GRAPHITE 9039, and GRAPHITE GA-17, available from Superior Graphite Co. The foregoing graphites exhibited an adsorption capacity for unsheared and sheared BAKER PETROLITE FLO[®] XS of about 0.01 wt% or more.

[0032] Preferred commercially available graphites for adsorbing unsheared BAKER PETROLITE FLO[®] XS and equivalents included GRAPHITE 2126, GRAPHITE 2139, GRAPHITE 3726, GRAPHITE 3739, GRAPHITE 5539, GRAPHITE 9039, and GRAPHITE GA-17. The foregoing graphites exhibited an adsorption capacity for unsheared BAKER PETROLITE FLO[®] XS of about 0.02 wt% or more. Preferred commercially available graphites for adsorbing sheared BAKER PETROLITE FLO[®]

20

XS and equivalents included GRAPHITE 2126, GRAPHITE 2139, GRAPHITE 3726, GRAPHITE 3739, GRAPHITE 9026, and GRAPHITE 9039. The foregoing graphites exhibited an adsorption capacity for sheared BAKER PETROLITE FLO[®] XS of about 0.018 wt% or more.

- 5 [0033] Even more preferred commercially available graphites for adsorbing unsheared BAKER PETROLITE FLO[®] XS and equivalents included GRAPHITE 2139, GRAPHITE 3726, GRAPHITE 3739, GRAPHITE 5539, GRAPHITE 9039, and GRAPHITE GA-17. The foregoing graphites exhibited an adsorption capacity for unsheared BAKER PETROLITE FLO[®] XS of about 0.03 wt% or more.
- 10 [0034] Most preferred graphites, particularly for adsorbing unsheared BAKER PETROLITE FLO[®] XS and equivalents thereof, include but are not necessarily limited to GRAPHITE 2139 and GRAPHITE 3739. The foregoing graphites exhibited an adsorption capacity for unsheared BAKER PETROLITE FLO[®] XS of about 0.04 wt% or more. Most preferred graphites, particularly for adsorbing sheared
- 15 BAKER PETROLITE FLO[®] XS and equivalents thereof, include but are not necessarily limited to GRAPHITE 3726 and GRAPHITE 3739. The foregoing graphites exhibited an adsorption capacity for sheared BAKER PETROLITE FLO[®] XS of about 0.025 wt% or more.

Removal of Drag Reducer Additive From Liquid Hydrocarbon Fuels

- 20 [0035] It may be desirable simply to subject all of a given fuel at a given storage or transport site to a DRA removal procedure. This would be particularly effective if all the DRA in use was removable by carbonaceous materials. In fact, it would even be

most preferable to use as DRA in fuels only materials known to be removable by carbonaceous materials. Or it may be desirable to test for the presence of DRA before incurring the expense of removal.

[0036] Once preferred carbonaceous materials, preferably graphite removal agents
5 have been identified, the activated carbon and/or graphite(s) may be used to remove the drag reducer additive from a given liquid hydrocarbon fuel, preferably motor gasoline or jet fuel, most preferably jet fuel. Alternately, a given sample of hydrocarbon fuel is analyzed for DRA by gel permeation chromatography (GPC).

[0037] When it is desired to remove DRA from a given fuel, one or more effective
10 carbonaceous materials, preferably selected activated carbons, more preferably graphite(s) are incorporated into a system for filtering the DRA/fuel mixture and for removing drag reducer additive from that mixture. The filter may be in any suitable form and may be installed in a variety of locations. Suitable locations include, but are not necessarily limited to a pipeline to a fuel terminal, a delivery system between a
15 fuel terminal and a tanker truck, a delivery system between two different tanker trucks, a delivery system from a tanker truck to a storage tank or to an engine, and, actually as a component of the engine, itself. In one embodiment, the filter comprises a component of a fuel delivery system from a tanker truck to a jet engine. The filter may be used in substantially any type of delivery system. In each method, the
20 graphites may or may not be heated. Heating removes any water, if any is adsorbed on the graphite.

[0038] Due to the difficulty in providing for incremental addition and agitation in

most commercial situations, it may be preferred to simply pass the liquid hydrocarbon fuel through a bed of the carbonaceous materials, preferably selected activated carbons, more preferably graphite(s), until the removal rate is so low that the carbonaceous materials must be replaced. In one embodiment, the filtering system provides for agitation of the DRA/fuel mixture as incremental additions of a given carbonaceous material, preferably a selected activated carbon(s), more preferably graphite(s) are added to the DRA/fuel mixture. This procedure is sometimes herein referred to as the "gradual addition and stirring" method or "Grad Add/Stir" method. This is a preferred method for more viscous hydrocarbon fuels such as jet fuel.

10 [0039] Civilian aircraft generally are serviced in hangers at airports. Military aircraft are serviced on the flight line, where a row of aircraft are parked away from a maintenance terminal and nearer to the runway, to be ready for mobilization. The distance from the flight line to the maintenance terminals may be as much as one mile. Servicing of aircraft, particularly military aircraft, typically is performed by shuttling service vehicles out to the flight line from the maintenance terminal where they perform the requisite service. Periodically, the servicing vehicles return to the maintenance terminal to themselves be refueled or for other servicing.

[0040] An example of how the system would be incorporated into a known fuel delivery system is described in U.S. Patent Application No. 10/124,974, filed on April 18, 2002, incorporated herein by reference. Briefly, the fuel delivery system comprises: (1) a tank of a refueling truck, (2) a transfer hose, (3) a bracket for storing the hose, (4) a manual pump, (5) a filter device, (6) a drain connected to the transfer

hose at one end, and (7) a shutoff valve. The filter device comprising the removal agent of the present application may be inserted either in series or in parallel with these one-way filters. Preferably, a filter device, such as a canister or cartridge of the removal agent(s), is placed downstream of the one-way filters in order to improve efficiency and longevity of the operation.

[0041] The application will be better understood with reference to the following examples, which are illustrative only:

EXAMPLE 1

10 [0042] The Grad Add/Stir method was used in this example. About 100 ml of jet fuel comprising about 8.36 ppm of FLO[®] XS - unsheared DRA (manufactured by Baker Petrolite) was stirred with a magnetic stir bar, to create a moderate vortex. Increments of about 0.02 to about 0.1 gram of a removal agent were placed in the agitating DRA/jet fuel mixture, while stirring, until a total of about 1.0 g had been
15 added. The stirring was continued for approximately two to three minutes. The sample was allowed to settle for about 5 minutes. The carbon was removed from the mixture by filtration with a Whatman 8 micron filter. The mixture was then tested for polymer concentration.

[0043] The polymer adsorption of removal agents increased with graphite as
20 compared to activated carbon. Table 1 (in Example 2) summarizes the % DRA adsorbed for both an activated carbon group (34 samples), and a graphite group (9 samples). FIG. 1 is a graphical representation of the effectiveness of graphites to

remove unsheared DRA from jet fuel. Table 1 further summarizes the % adsorption capacity for both an activated carbon group, and a graphite group. For unsheared DRA, the % adsorption capacity of a given carbon = $(0.000678) * (\% \text{ polymer removed})$. This formula is derived using the initial polymer concentration, volume of
5 DRA/fuel used in the experiment, weight of carbon used in the experiment, and density of the DRA/fuel used.

[0044] As shown in FIG. 1, graphites are more effective at removing unsheared polymer than activated carbons. The highest removal effectiveness of the 43 removal agents tested was 58.20 % (with "Graphite 3739"), provided by Superior Graphite
10 Company, which corresponds to a polymer adsorption capacity of 0.04 % (wt).

EXAMPLE 2

[0045] The Quick Add/Stir method was used in this example. About 100 ml of jet fuel comprising about 8.36 ppm of FLO[®] XS - unsheared DRA (manufactured by Baker Petrolite) was stirred with a magnetic stir bar, to create a moderate vortex.
15 Once the removal agent had cooled, about 1.0 g of the removal agent was placed in the agitating DRA/jet fuel mixture, while stirring. The stirring was continued for approximately two to three minutes. The sample was allowed to settle for about 5 minutes. The carbon was removed from the mixture by filtration with a Whatman 8 micron filter. The mixture was then tested for polymer concentration.

20 [0046] The polymer adsorption of removal agents increased with graphite as compared to activated carbon. Table 1 summarizes the % DRA adsorbed for both an activated carbon group (7 samples), and a graphite group (1 sample). The highest

removal effectiveness of the 8 removal agents tested was 49.90 % (with "Graphite 3739"), which corresponds to a polymer adsorption capacity of 0.034 %(wt). FIG. 2 compares the effectiveness of removing DRA using the Grad Add/Stir and Quick Add/Stir methods for the 8 commonly tested removal agents. The Grad Add/Stir method gave superior results in 5 of the 8 carbons tested. The Quick Add/Stir method gave superior results in 3 of the 8 carbons tested. FIG. 2 further demonstrates that graphite is more effective at DRA removal than activated carbon, regardless of the method employed.

[0047] The removal agents listed in Table 1 are derived from numerous sources, including wood, coconut and other shells, peat, bituminous coal, lignite coal, and anthracite coal. Each carbon source material differs in surface area, pore size, density, and strength characteristics.

Table 1

Sample	% Polymer Removal (Grad Add/Stir Method) FLO® XS - Unsheared	% Adsorption Capacity (Grad Add/Stir Method) FLO® XS - Unsheared	% Polymer Removal (Quick Add/Stir Method) FLO® XS - Unsheared	% Adsorption Capacity (Quick Add/Stir Method) FLO® XS - Unsheared
Activated Carbon				
(1)Calgon ADP	22.40	0.015		
(2)Calgon APL	2.43	0.002		
(3)Calgon BL	6.44	0.004		
(4)Calgon C	6.68	0.005		
(5)Calgon Colorsorb	23.70	0.016	4.78	0.003
(6)Calgon RB	4.13	0.003	9.81	0.070
(7)Calgon RC	0.00	0.00		
(8)Calgon WPH	6.93	0.005		
(9)Calgon WPL	4.98	0.003		
(10)Calgon WPX	29.20	0.020	5.02	0.003
(11)Darco G-60	9.60	0.007		
(12)Darco S-51	9.48	0.006		
(13)Norit A Supra	38.80	0.026	25.20	0.020

(14)Norit CA1	37.10	0.025	2.27	0.002
(15)Norit CASPF	18.30	0.012		
(16)Norit E Supra	0.00	0.00		
(17)Norit FGD	47.00	0.032	42.60	0.030
(18)Norit HDB	36.10	0.025		
(19)Norit KB	16.50	0.011	15.70	0.011
(20)Norit PAC200	0.00	0.00		
(21)Norit SX Plus	0.00	0.00		
(22)Norit SX 4	2.19	0.002		
(23)Aquacarb 1230C	20.20	0.014		
(24)Aquacarb 1230CAW	4.98	0.003		
(25)Aquacarb 1240	4.01	0.003		
(26)Aquacarb 1240AW	3.77	0.003		
(27)Aquapac 800	14.00	0.010		
(28)Bevcarb 800	0.00	0.00		
(29)Ultracarb 1240	14.60	0.010		
(30)Chemcarb C25	3.65	0.003		
(31)PAC 2000-600	17.90	0.012		
(32)PAC 2000-900	7.90	0.005		
(33)SX0 Powder	35.80	0.024		
(34)Carbon 5565	33.40	0.023		
Graphite				
(35)Graphite 2126	29.30	0.020		
(36)Graphite 2139	52.60	0.040		
(37)Graphite 3726	37.20	0.030		
(38)Graphite 3739	58.20	0.040	49.90	0.034
(39)Graphite 5526	14.30	0.010		
(40)Graphite 5539	43.60	0.030		
(41)Graphite 9026	20.40	0.014		
(42)Graphite 9039	37.90	0.030		
(43)Graphite GA-17	43.50	0.030		

[0048] Based on the foregoing, a gradual or incremental add/stir procedure is more efficient in removing DRA from more viscous hydrocarbon fuels, such as jet fuel.

EXAMPLE 3

5 [0049] The Grad Add/Stir method was used in this example. About 100 ml of jet fuel comprising about 8.36 ppm of FLO[®] XS - sheared DRA (manufactured by Baker Petrolite) was stirred with a magnetic stir bar, to create a moderate vortex. Increments of about 0.02 to about 0.1 gram of a removal agent were placed in the agitating DRA/jet fuel mixture, while stirring, until a total of about 1.0 g had been added. The
10 stirring was continued for approximately two to three minutes. The sample was allowed to settle for about 5 minutes. The carbon was removed from the mixture by filtration with a Whatman 8 micron filter. The mixture was then tested for polymer concentration.

[0050] The polymer adsorption of removal agents increased with graphite as
15 compared to activated carbon. Table 2 (in Example 4) summarizes the % DRA adsorbed for both an activated carbon group (34 samples), and a graphite group (9 samples). FIG. 3 is a graphical representation of the effectiveness of graphites to remove sheared DRA from jet fuel. Table 2 further summarizes the % adsorption capacity for both an activated carbon group, and a graphite group. For sheared DRA,
20 the % adsorption capacity of a given carbon = $(0.000857) * (\% \text{ polymer removed})$.
This formula is derived using the initial polymer concentration, volume of DRA/fuel used in the experiment, weight of carbon used in the experiment, and density of the

DRA/fuel used.

[0051] As shown in FIG. 3, graphites are more effective at removing sheared polymer than activated carbons. The highest removal effectiveness of the 43 removal agents tested was 36.30% (with "Graphite 3739"), which corresponds to a polymer
5 adsorption capacity of 0.031 %(wt).

EXAMPLE 4

[0052] The Quick Add/Stir method was used in this example. About 100 ml of jet fuel comprising about 8.36 ppm of FLO[®] XS - sheared DRA was stirred with a magnetic stir bar, to create a moderate vortex. Once the removal agent had cooled,
10 about 1.0 g of the removal agent was placed in the agitating DRA/jet fuel mixture, while stirring. The stirring was continued for approximately two to three minutes. The sample was allowed to settle for about 5 minutes. The carbon was removed from the mixture by filtration with a Whatman 8 micron filter. The mixture was then tested for polymer concentration.

15 [0053] The polymer adsorption of removal agents increased with graphite as compared to activated carbon. Table 2 summarizes the % DRA adsorbed for both an activated carbon group (7 samples), and a graphite group (1 sample). The highest removal effectiveness of the 8 removal agents tested was 42.10 % (with "Graphite 3739"), which corresponds to a polymer adsorption capacity of 0.040 %(wt). FIG. 4
20 compares the effectiveness of removing DRA using the Grad Add/Stir and Quick Add/Stir methods for the 8 commonly tested removal agents. The Grad Add/Stir method gave superior results in 5 of the 8 carbons tested. The Quick Add/Stir method

gave superior results in 3 of the 8 carbons tested. FIG. 4 further demonstrates that graphite is more effective at DRA removal than activated carbon, regardless of the method employed.

[0054] FIG. 5 demonstrates the effectiveness of the removal agents to remove both sheared and unsheared FLO[®] XS from jet fuel. All removal agents perform better removing unsheared polymer over sheared polymer. Only the most effective removal agents at removing unsheared polymer were effective at removing sheared polymer.

Table 2

Sample	% Polymer Removal (Grad Add/Stir Method) FLO [®] XS - Sheared	% Adsorption Capacity (Grad Add/Stir Method) FLO [®] XS - Sheared	% Polymer Removal (Quick Add/Stir Method) FLO [®] XS - Sheared	% Adsorption Capacity (Quick Add/Stir Method) FLO [®] XS - Sheared
Activated Carbon				
(1)Calgon ADP	00.00	00.00		
(2)Calgon APL	1.900	0.002		
(3)Calgon BL	00.95	0.001		
(4)Calgon C	5.900	0.010		
(5)Calgon Colorsorb	5.330	0.005	6.100	0.010
(6)Calgon RB	4.950	0.004	00.00	00.00
(7)Calgon RC	3.810	0.003		
(8)Calgon WPH	6.950	0.006		
(9)Calgon WPL	4.760	0.004		
(10)Calgon WPX	4.860	0.004	5.81	0.005
(11)Darco G-60	00.00	00.00		
(12)Darco S-51	4.860	0.004		
(13)Norit A Supra	2.860	0.003	0.95	0.001
(14)Norit CA1	0.950	0.001	00.00	00.00
(15)Norit CASPF	00.00	00.00		
(16)Norit E Supra	00.00	00.00		
(17)Norit FGD	32.40	0.030	26.70	0.023
(18)Norit HDB	22.50	0.020		
(19)Norit KB	1.900	0.002	0.95	0.001
(20)Norit PAC200	3.810	0.003		

(21)Norit SX Plus	00.00	00.00		
(22)Norit SX 4	3.810	0.003		
(23)Aquacarb 1230C	14.00	0.012		
(24)Aquacarb 1230CAW	00.00	00.00		
(25)Aquacarb 1240	1.900	0.002		
(26)Aquacarb 1240AW	00.00	00.00		
(27)Aquapac 800	10.00	0.010		
(28)Bevcarb 800	4.950	0.004		
(29)Ultracarb 1240	7.430	0.010		
(30)Chemcarb C25	1.900	0.002		
(31)PAC 2000-600	4.950	0.004		
(32)PAC 2000-900	5.330	0.005		
(33)SX0 Powder	29.30	0.030		
(34)Carbon 5565	23.60	0.020		
Graphite				
(35)Graphite 2126	19.30	0.020		
(36)Graphite 2139	25.00	0.021		
(37)Graphite 3726	33.40	0.030		
(38)Graphite 3739	36.30	0.031	42.10	0.040
(39)Graphite 5526	8.670	0.007		
(40)Graphite 5539	20.40	0.017		
(41)Graphite 9026	18.10	0.020		
(42)Graphite 9039	19.20	0.020		
(43)Graphite GA-17	7.990	0.007		

[0055] Persons of ordinary skill in the art will recognize that many modifications may be made to the present application without departing from the spirit and scope of the present application. The embodiment described herein is meant to be illustrative

5 only and should not be taken as limiting the application.

We claim:

- 1 1. A method of removing drag reducer additive from a liquid
2 hydrocarbon fuel, said method comprising:
3 providing a contaminated liquid hydrocarbon fuel comprising a concentration
4 of a drag reducer additive (DRA);
5 contacting said contaminated liquid hydrocarbon fuel with a quantity of one or
6 more graphite effective to substantially reduce said concentration of
7 DRA, said contacting occurring under conditions effective to produce a
8 clean liquid hydrocarbon fuel.
- 1 2. The method of claim 1 wherein said graphite comprises a graphite
2 powder.
- 1 3. The method of claim 1 wherein said graphite comprises a particulate
2 having an average diameter of from about 1 micron to about 100 microns.
- 1 4. The method of claim 1 wherein said graphite comprises an adsorption
2 capacity for said DRA of about 0.01 wt.% or more at from about 8 to about 12 ppm
3 DRA concentration and at about 1g activated carbon/100 ml fuel, said DRA being
4 selected from the group consisting of sheared and unsheared DRA.
- 1 5. The method of claim 1 wherein said graphite comprises an adsorption
2 capacity for DRA of about 0.02 wt.% or more at from about 8 to about 12 ppm DRA
3 concentration and at about 1g activated carbon/100 ml fuel, said DRA being selected
4 from the group consisting of sheared and unsheared DRA.
- 1 6. The method of claim 1 wherein said graphite comprises an adsorption

2 capacity for sheared DRA of about 0.025 wt.% or more at from about 8 to about 12
3 ppm DRA concentration and at about 1g activated carbon/100 ml fuel.

1 7. The method of claim 1 wherein said graphite comprises an adsorption
2 capacity for unsheared DRA of about 0.04 wt.% or more at from about 8 to about 12
3 ppm DRA concentration and at about 1g activated carbon/100 ml fuel.

1 8. The method of claim 1 wherein said graphite produces a % polymer
2 removal for unsheared DRA of about 25% or more at from about 8 to about 12 ppm
3 DRA concentration and at about 1g activated carbon/100 ml fuel.

1 9. The method of claim 1 wherein said graphite produces a % polymer
2 removal for DRA of about 30% or more at from about 8 to about 12 ppm DRA
3 concentration and at about 1g activated carbon/100 ml fuel, said DRA being selected
4 from the group consisting of sheared and unsheared DRA.

1 10. The method of claim 1 wherein said graphite produces a % polymer
2 removal for sheared DRA of about 35% or more at from about 8 to about 12 ppm
3 DRA concentration and at about 1g activated carbon/100 ml fuel.

1 11. The method of claim 1 wherein said graphite produces a % DRA
2 removal for unsheared DRA of about 40% or more at about 1g activated carbon/100
3 ml fuel.

1 12. The method of claim 1 wherein said graphite produces a % DRA
2 removal for unsheared DRA of about 50% or more at from about 8 to about 12 ppm
3 DRA concentration and at about 1g activated carbon/100 ml fuel.

1 13. The method of claim 1 wherein said graphite is selected from the group

2 consisting of GRAPHITE 2126, GRAPHITE 2139, GRAPHITE 3726, GRAPHITE
3 3739, GRAPHITE 5526, GRAPHITE 5539, GRAPHITE 9026, GRAPHITE 9039,
4 and GRAPHITE GA-17.

1 14. The method of claim 1 wherein said graphite is selected from the group
2 consisting of GRAPHITE 2126, GRAPHITE 2139, GRAPHITE 3726, GRAPHITE
3 3739, GRAPHITE 5539, GRAPHITE 9039, and GRAPHITE GA-17.

1 15. The method of claim 1 wherein said graphite is selected from the group
2 consisting of GRAPHITE 2139, GRAPHITE 3726, GRAPHITE 3739, GRAPHITE
3 5539, GRAPHITE 9039, and GRAPHITE GA-17.

1 16. The method of claim 1 wherein said graphite is selected from the group
2 consisting of GRAPHITE 2139 and GRAPHITE 3739.

1 17. A method of removing drag reducer additive from a liquid
2 hydrocarbon fuel, said method comprising:
3 providing a contaminated liquid hydrocarbon fuel comprising a concentration
4 of a drag reducer additive (DRA);
5 contacting said contaminated liquid hydrocarbon fuel with a quantity of one or
6 more activated carbon effective to having an adsorption capacity for
7 DRA of about 0.014 or more at from about 8 to about 12 ppm DRA
8 concentration and at about 1g activated carbon/100 ml fuel, said DRA
9 being sheared or unsheared, said contacting occurring under conditions
10 effective to produce a clean liquid hydrocarbon fuel.

1 18. The method of claim 17 wherein said activated carbon has an

2 adsorption capacity for sheared DRA of 0.018% or more.

1 19. The method of claim 17 wherein said activated carbon has an
2 adsorption capacity for DRA of about 0.020% or more, said DRA being selected from
3 the group consisting of sheared and unsheared DRA.

1 20. The method of claim 17 wherein said activated carbon has an
2 adsorption capacity for DRA of 0.025%, said DRA being selected from the group
3 consisting of sheared and unsheared DRA.

1 21. The method of claim 17 wherein said activated carbon has an
2 adsorption capacity for DRA of about 0.030%, said DRA being selected from the
3 group consisting of sheared and unsheared DRA.

1 22. The method of claim 17 wherein said activated carbon comprises an
2 activated carbon powder.

1 23. The method of claim 17 wherein said activated carbon comprises a
2 particulate having an average diameter of from about 1 micron to about 100 microns.

1 24. The method of claim 17 wherein said activated carbon produces a %
2 polymer removal for DRA of about 20% or more at from about 8 to about 12 ppm
3 DRA concentration and at about 1g activated carbon/100 ml fuel, said DRA being
4 selected from the group consisting of sheared and unsheared DRA.

1 25. The method of claim 17 wherein said activated carbon produces a %
2 polymer removal for unsheared DRA of about 30% or more at from about 8 to about
3 12 ppm DRA concentration and at about 1g activated carbon/100 ml fuel, said DRA
4 being selected from the group consisting of sheared and unsheared DRA.

1 26. The method of claim 17 wherein said activated carbon produces a %
2 polymer removal for unsheared DRA of about 40% or more at from about 8 to about
3 12 ppm DRA concentration and at about 1g activated carbon/100 ml fuel.

1 27. The method of claim 17 wherein said activated carbon is selected from
2 the group consisting of CALGON ADP, CALGON COLORSORB, CALGON WPX,
3 NORIT A SUPRA, NORIT CA 1, NORITN FGD, NORIT HDB, SXO POWDER,
4 and CARBON 5565.

1 28. The method of claim 17 wherein said activated carbon is selected from
2 the group consisting of CALGON WPX, NORIT A SUPRA, NORIT CA1, NORIT
3 FGD, NORIT HDB, SXO POWDER and CARBON 5565.

1 29. The method of claim 17 wherein said activated carbon is selected from
2 the group consisting of NORIT A SUPRA, NORIT CA1, NORIT FGD, and NORIT
3 HDB.

1 30. A method of removing drag reducer additive from a liquid hydrocarbon
2 fuel, said method comprising:

3 providing a contaminated liquid hydrocarbon fuel comprising a concentration
4 of a drag reducer additive (DRA), said DRA comprising
5 polyalphaolefin produced by solution polymerization;
6 contacting said contaminated liquid hydrocarbon fuel with a quantity of
7 removal agent under conditions effective to produce a clean liquid
8 hydrocarbon fuel, said removal agent being selected from the group
9 consisting of one or more graphite and one or more activated carbon

10 having an adsorption capacity for said DRA of about 0.014 or more at
11 from about 8 to about 12 ppm DRA concentration and at about 1g
12 activated carbon/100 ml fuel.

1 31. The method of claim 30 wherein said graphite comprises an adsorption
2 capacity for DRA of about 0.02 wt.% or more at from about 8 to about 12 ppm DRA
3 concentration and at about 1g activated carbon/100 ml fuel, said DRA being selected
4 from the group consisting of sheared and unsheared DRA.

1 32. The method of claim 30 wherein said graphite comprises an
2 adsorption capacity for unsheared DRA of about 0.04 wt.% or more at from about 8 to
3 about 12 ppm DRA concentration and at about 1g activated carbon/100 ml fuel.

1 33. The method of claim 30 wherein said graphite produces a % polymer
2 removal for unsheared DRA of about 25% or more at from about 8 to about 12 ppm
3 DRA concentration and at about 1g activated carbon/100 ml fuel.

1 34. The method of claim 30 wherein said graphite produces a % polymer
2 removal for DRA of about 30% or more at from about 8 to about 12 ppm DRA
3 concentration and at about 1g activated carbon/100 ml fuel, said DRA being selected
4 from the group consisting of sheared and unsheared DRA.

1 35. The method of claim 30 wherein said graphite produces a % DRA
2 removal for unsheared DRA of about 40% or more at about 1g activated carbon/100
3 ml fuel.

1 36. The method of claim 30 wherein said graphite produces a % DRA
2 removal for unsheared DRA of about 50% or more at from about 8 to about 12 ppm

3 DRA concentration and at about 1g activated carbon/100 ml fuel.

1 37. The method of claim 30 wherein said removal agent comprises graphite
2 selected from the group consisting of GRAPHITE 2126, GRAPHITE 2139,
3 GRAPHITE 3726, GRAPHITE 3739, GRAPHITE 5526, GRAPHITE 5539,
4 GRAPHITE 9026, GRAPHITE 9039, and GRAPHITE GA-17.

1 38. The method of claim 30 wherein said removal agent comprises
2 graphite selected from the group consisting of GRAPHITE 2126, GRAPHITE 2139,
3 GRAPHITE 3726, GRAPHITE 3739, GRAPHITE 5539, GRAPHITE 9039, and
4 GRAPHITE GA-17.

1 39. The method of claim 30 wherein said removal agent comprises
2 graphite selected from the group consisting of GRAPHITE 2139, GRAPHITE 3726,
3 GRAPHITE 3739, GRAPHITE 5539, GRAPHITE 9039, and GRAPHITE GA-17.

1 40. The method of claim 30 wherein said removal agent comprises
2 graphite selected from the group consisting of GRAPHITE 2139 and GRAPHITE
3 3739.

1 41. The method of claim 30 wherein said removal agent is activated carbon
2 selected from the group consisting of CALGON ADP, CALGON COLORSORB,
3 CALGON WPX, NORIT A SUPRA, NORIT CA 1, NORITN FGD, NORIT HDB,
4 SXO POWDER, and CARBON 5565.

1 42. The method of claim 30 wherein said removal agent is activated carbon
2 selected from the group consisting of CALGON WPX, NORIT A SUPRA, NORIT
3 CA1, NORIT FGD, NORIT HDB, SXO POWDER and CARBON 5565.

1 43. The method of claim 30 wherein said removal agent is activated carbon
2 selected from the group consisting of NORIT A SUPRA, NORIT CA1, NORIT FGD,
3 and NORIT HDB.

1 44. A method of removing drag reducer additive from a liquid hydrocarbon
2 fuel, said method comprising:
3 providing a contaminated liquid hydrocarbon fuel comprising a concentration
4 of a drag reducer additive (DRA) comprising polymerized linear alpha
5 olefin (LAO) monomers having from about 6 to about 12 carbon
6 atoms, wherein said LAO monomers comprise two different LAO's
7 which differ in number of carbon atoms by 6;
8 contacting said contaminated liquid hydrocarbon fuel with a quantity of a
9 removal agent under conditions effective to produce a clean liquid
10 hydrocarbon fuel, said removal agent being selected from the group
11 consisting of one or more graphite and one or more activated carbon
12 having an adsorption capacity for said DRA of about 0.014 or more at
13 from about 8 to about 12 ppm DRA concentration and at about 1g
14 activated carbon/100 ml fuel.

1 45. The method of claim 44 wherein said graphite comprises an adsorption
2 capacity for DRA of about 0.02 wt.% or more at from about 8 to about 12 ppm DRA
3 concentration and at about 1g activated carbon/100 ml fuel, said DRA being selected
4 from the group consisting of sheared and unsheared DRA.

1 46. The method of claim 44 wherein said graphite comprises an adsorption

2 capacity for unsheared DRA of about 0.04 wt. % or more at from about 8 to about 12
3 ppm DRA concentration and at about 1g activated carbon/100 ml fuel.

1 47. The method of claim 44 wherein said graphite produces a % polymer
2 removal for unsheared DRA of about 25% or more at from about 8 to about 12 ppm
3 DRA concentration and at about 1g activated carbon/100 ml fuel.

1 48. The method of claim 44 wherein said graphite produces a % polymer
2 removal for DRA of about 30% or more at from about 8 to about 12 ppm DRA
3 concentration and at about 1g activated carbon/100 ml fuel, said DRA being selected
4 from the group consisting of sheared and unsheared DRA.

1 49. The method of claim 44 wherein said graphite produces a % DRA
2 removal for unsheared DRA of about 40% or more at about 1g activated carbon/100
3 ml fuel.

1 50. The method of claim 44 wherein said graphite produces a % DRA
2 removal for unsheared DRA of about 50% or more at from about 8 to about 12 ppm
3 DRA concentration and at about 1g activated carbon/100 ml fuel.

1 51. The method of claim 44 wherein said removal agent comprises
2 graphite selected from the group consisting of GRAPHITE 2126, GRAPHITE 2139,
3 GRAPHITE 3726, GRAPHITE 3739, GRAPHITE 5526, GRAPHITE 5539,
4 GRAPHITE 9026, GRAPHITE 9039, and GRAPHITE GA-17.

1 52. The method of claim 44 wherein said removal agent comprises
2 graphite selected from the group consisting of GRAPHITE 2126, GRAPHITE 2139,
3 GRAPHITE 3726, GRAPHITE 3739, GRAPHITE 5539, GRAPHITE 9039, and

4 GRAPHITE GA-17.

1 53. The method of claim 44 wherein said removal agent comprises
2 graphite selected from the group consisting of GRAPHITE 2139, GRAPHITE 3726,
3 GRAPHITE 3739, GRAPHITE 5539, GRAPHITE 9039, and GRAPHITE GA-17.

1 54. The method of claim 44 wherein said removal agent comprises
2 graphite selected from the group consisting of GRAPHITE 2139 and GRAPHITE
3 3739.

1 55. The method of claim 44 wherein said removal agent is activated carbon
2 selected from the group consisting of CALGON ADP, CALGON COLORSORB,
3 CALGON WPX, NORIT A SUPRA, NORIT CA 1, NORITN FGD, NORIT HDB,
4 SXO POWDER, and CARBON 5565.

1 56. The method of claim 44 wherein said removal agent is activated carbon
2 selected from the group consisting of CALGON WPX, NORIT A SUPRA, NORIT
3 CA1, NORIT FGD, NORIT HDB, SXO POWDER and CARBON 5565.

1 57. The method of claim 44 wherein said removal agent is activated carbon
2 selected from the group consisting of NORIT A SUPRA, NORIT CA1, NORIT FGD,
3 and NORIT HDB.

1 58. A method of removing drag reducer additive from a liquid hydrocarbon
2 fuel, said method comprising:

3 providing a contaminated liquid hydrocarbon fuel comprising a concentration
4 of a drag reducer additive (DRA) comprising polymerized linear alpha
5 olefin (LAO) monomers having from about 2 to about 40 carbon

6 atoms;

7 contacting said contaminated liquid hydrocarbon fuel with a quantity of a
8 removal agent under conditions effective to produce a clean liquid
9 hydrocarbon fuel, said removal agent being selected from the group
10 consisting of one or more graphite and one or more activated carbon
11 having an adsorption capacity for said DRA of about 0.014 or more at
12 from about 8 to about 12 ppm DRA concentration and at about 1g
13 activated carbon/100 ml fuel.

1 59. The method of claim 58 wherein said LAO monomers have from about
2 2 to about 30 carbon atoms.

3 60. The method of claim 58 wherein said LAO monomers have from about
4 4 to about 20 carbon atoms.

5 61. The method of claim 58 wherein said LAO monomers have from about
6 6 to about 12 carbon atoms.

7 62. The method of claim 58 wherein said graphite comprises an adsorption
8 capacity for DRA of about 0.02 wt.% or more at from about 8 to about 12 ppm DRA
9 concentration and at about 1g activated carbon/100 ml fuel, said DRA being selected
10 from the group consisting of sheared and unsheared DRA.

1 63. The method of claim 58 wherein said graphite comprises an
2 adsorption capacity for unsheared DRA of about 0.04 wt.% or more at from about 8 to
3 about 12 ppm DRA concentration and at about 1g activated carbon/100 ml fuel.

1 64. The method of claim 58 wherein said graphite produces a % polymer

2 removal for unsheared DRA of about 25% or more at from about 8 to about 12 ppm
3 DRA concentration and at about 1g activated carbon/100 ml fuel.

1 65. The method of claim 58 wherein said graphite produces a % polymer
2 removal for DRA of about 30% or more at from about 8 to about 12 ppm DRA
3 concentration and at about 1g activated carbon/100 ml fuel, said DRA being selected
4 from the group consisting of sheared and unsheared DRA.

1 66. The method of claim 58 wherein said graphite produces a % DRA
2 removal for unsheared DRA of about 40% or more at about 1g activated carbon/100
3 ml fuel.

1 67. The method of claim 58 wherein said graphite produces a % DRA
2 removal for unsheared DRA of about 50% or more at from about 8 to about 12 ppm
3 DRA concentration and at about 1g activated carbon/100 ml fuel.

1 68. The method of claim 58 wherein said graphite comprises an adsorption
2 capacity for DRA of about 0.02 wt.% or more at from about 8 to about 12 ppm DRA
3 concentration and at about 1g activated carbon/100 ml fuel, said DRA being selected
4 from the group consisting of sheared and unsheared DRA.

1 69. The method of claim 58 wherein said graphite produces a % polymer
2 removal for DRA of about 30% or more at from about 8 to about 12 ppm DRA
3 concentration and at about 1g activated carbon/100 ml fuel, said DRA being selected
4 from the group consisting of sheared and unsheared DRA.

1 70. The method of claim 58 wherein said removal agent comprises
2 graphite selected from the group consisting of GRAPHITE 2126, GRAPHITE 2139,

3 GRAPHITE 3726, GRAPHITE 3739, GRAPHITE 5526, GRAPHITE 5539,
4 GRAPHITE 9026, GRAPHITE 9039, and GRAPHITE GA-17.

1 71. The method of claim 58 wherein said removal agent comprises
2 graphite selected from the group consisting of GRAPHITE 2126, GRAPHITE 2139,
3 GRAPHITE 3726, GRAPHITE 3739, GRAPHITE 5539, GRAPHITE 9039, and
4 GRAPHITE GA-17.

1 72. The method of claim 58 wherein said removal agent comprises
2 graphite selected from the group consisting of GRAPHITE 2139, GRAPHITE 3726,
3 GRAPHITE 3739, GRAPHITE 5539, GRAPHITE 9039, and GRAPHITE GA-17.

1 73. The method of claim 58 wherein said removal agent comprises
2 graphite selected from the group consisting of GRAPHITE 2139 and GRAPHITE
3 3739.

1 74. The method of claim 58 wherein said removal agent is activated carbon
2 selected from the group consisting of CALGON ADP, CALGON COLORSORB,
3 CALGON WPX, NORIT A SUPRA, NORIT CA 1, NORITN FGD, NORIT HDB,
4 SXO POWDER, and CARBON 5565.

1 75. The method of claim 58 wherein said removal agent is activated carbon
2 selected from the group consisting of CALGON WPX, NORIT A SUPRA, NORIT
3 CA1, NORIT FGD, NORIT HDB, SXO POWDER and CARBON 5565.

1 76. The method of claim 58 wherein said removal agent is activated carbon
2 selected from the group consisting of NORIT A SUPRA, NORIT CA1, NORIT FGD,
3 and NORIT HDB.

- 1 77. The method of claim 58 wherein said removal agent comprises
2 graphite selected from the group consisting of GRAPHITE 2126, GRAPHITE 2139,
3 GRAPHITE 3726, GRAPHITE 3739, GRAPHITE 5526, GRAPHITE 5539,
4 GRAPHITE 9026, GRAPHITE 9039, and GRAPHITE GA-17.
- 1 78. The method of claim 58 wherein said removal agent comprises
2 graphite selected from the group consisting of GRAPHITE 2139 and GRAPHITE
3 3739.
- 1 79. The method of claim 58 wherein said removal agent is activated carbon
2 selected from the group consisting of CALGON ADP, CALGON COLORSORB,
3 CALGON WPX, NORIT A SUPRA, NORIT CA 1, NORITN FGD, NORIT HDB,
4 SXO POWDER, and CARBON 5565.
- 1 80. The method of claim 58 wherein said removal agent is activated carbon
2 selected from the group consisting of NORIT A SUPRA, NORIT CA1, NORIT FGD,
3 and NORIT HDB.
- 4 81. The method of any of claims 1 and 2-80 wherein the liquid
5 hydrocarbon fuel has a boiling range of from about 150 °F to about 750 °F.
- 1 82. The method of any of claims 1 and 2-80 wherein said liquid
2 hydrocarbon fuel is selected from the group consisting of liquefied natural gas (LNG),
3 liquefied petroleum gas (LPG), motor gasoline, aviation gasoline, distillate fuels such
4 as diesel fuel and home heating oil, kerosene, jet fuel, No. 2 oil, residual fuel, No. 6
5 fuel, or bunker fuel.
- 1 83. The method of claims 1 and 2-80 wherein the liquid hydrocarbon fuel

2 is selected from the group consisting of diesel fuel, jet fuel, aviation gasoline, and
3 motor gasoline.

1 84. The method of claims 1 and 2-80 wherein the liquid hydrocarbon fuel
2 is jet fuel.

1 85. The method of any of claims 1 and 2-80 wherein the DRA is FLO[®] XS.

1 86. The method of claim 84 wherein the DRA is FLO[®] XS.

1 87. The method of any of claims 1 and 2-80 wherein said conditions
2 comprise adding said quantity of said one or more effective graphites to said
3 contaminated liquid hydrocarbon fuel in increments with agitation.

1 88. The method of claim 81 wherein said conditions comprise adding said
2 quantity of said one or more effective graphites to said contaminated liquid
3 hydrocarbon fuel in increments with agitation.

1 89. The method of claim 82 wherein said conditions comprise adding said
2 quantity of said one or more effective graphites to said contaminated liquid
3 hydrocarbon fuel in increments with agitation.

1 90. The method of claim 83 wherein said conditions comprise adding said
2 quantity of said one or more effective graphites to said contaminated liquid
3 hydrocarbon fuel in increments with agitation.

1 91. The method of claim 84 wherein said conditions comprise adding said
2 quantity of said one or more effective graphites to said contaminated liquid
3 hydrocarbon fuel in increments with agitation.

1 92. The method of claim 85 wherein said conditions comprise adding said

2 quantity of said one or more effective graphites to said contaminated liquid
3 hydrocarbon fuel in increments with agitation.

1 93. The method of claim 86 wherein said conditions comprise adding said
2 quantity of said one or more effective graphites to said contaminated liquid
3 hydrocarbon fuel in increments with agitation.

1 94. The method of claim 87 wherein said conditions comprise adding said
2 quantity of said one or more effective graphites to said contaminated liquid
3 hydrocarbon fuel in increments with agitation.

1 95. A method for selecting a drag reducer additive for a liquid hydrocarbon
2 fuel comprising:
3 providing one or more samples of liquid hydrocarbon fuel comprising a single
4 candidate DRA; and,
5 determining whether said single candidate DRA is removed from said liquid
6 hydrocarbon fuel by a removal agent selected from the group
7 consisting of a graphite and an activated carbon having an
8 adsorption capacity for DRA of about 0.014 or more at from about 8 to
9 about 12 ppm DRA concentration and at about 1g activated carbon/100
10 ml fuel; and,
11 selecting as said DRA only candidates which are removed from said liquid
12 hydrocarbon fuel by said removal agent.
1 96. The method of claim 95 wherein said liquid hydrocarbon fuel in said
2 samples is jet fuel.

1 97. The method of claims 95 and 96 wherein said removal agent is
2 activated carbon selected from the group consisting of CALGON ADP, CALGON
3 COLORSORB, CALGON WPX, NORIT A SUPRA, NORIT CA 1, NORITN FGD,
4 NORIT HDB, SXO POWDER, and CARBON 5565.

1 98. The method of claims 95 and 96 wherein said removal agent is
2 activated carbon selected from the group consisting of CALGON WPX, NORIT A
3 SUPRA, NORIT CA1, NORIT FGD, NORIT HDB, SXO POWDER and CARBON
4 5565.

1 99. The method of claims 95 and 96 wherein said removal agent is
2 activated carbon selected from the group consisting of NORIT A SUPRA, NORIT
3 CA1, NORIT FGD, and NORIT HDB.

4 100. The method of claims 95 and 96 wherein said removal agent is graphite
5 selected from the group consisting of GRAPHITE 2126, GRAPHITE 2139,
6 GRAPHITE 3726, GRAPHITE 3739, GRAPHITE 5526, GRAPHITE 5539,
7 GRAPHITE 9026, GRAPHITE 9039, and GRAPHITE GA-17.

1 101. The method of claims 95 and 96 wherein said removal agent is graphite
2 selected from the group consisting of GRAPHITE 2126, GRAPHITE 2139,
3 GRAPHITE 3726, GRAPHITE 3739, GRAPHITE 5539, GRAPHITE 9039, and
4 GRAPHITE GA-17.

1 102. The method of claims 95 and 96 wherein said removal agent is graphite
2 selected from the group consisting of GRAPHITE 2139, GRAPHITE 3726,
3 GRAPHITE 3739, GRAPHITE 5539, GRAPHITE 9039, and GRAPHITE GA-17.

1 103. The method of claims 95 and 96 wherein said removal agent comprises
2 GRAPHITE 2139

3 104. The method of claims 95 and 96 wherein said removal agent comprises
4 and GRAPHITE 3739.

1 105. A removal agent for removing drag reducer from a liquid hydrocarbon
2 fuel, the removal agent comprising activated carbon selected from the group
3 consisting of CALGON ADP, CALGON COLORSORB, CALGON WPX, NORIT A
4 SUPRA, NORIT CA 1, NORITN FGD, NORIT HDB, SXO POWDER, and
5 CARBON 5565.

1 106. A removal agent for removing drag reducer from a liquid hydrocarbon
2 fuel, the removal agent comprising activated carbon selected from the group
3 consisting of CALGON WPX, NORIT A SUPRA, NORIT CA1, NORIT FGD,
4 NORIT HDB, SXO POWDER and CARBON 5565.

1 107. A removal agent for removing drag reducer from a liquid hydrocarbon
2 fuel, the removal agent comprising activated carbon selected from the group
3 consisting of NORIT A SUPRA, NORIT CA1, NORIT FGD, and NORIT HDB.

4 108. A removal agent for removing drag reducer from a liquid hydrocarbon
5 fuel, the removal agent comprising graphite selected from the group consisting of
6 GRAPHITE 2126, GRAPHITE 2139, GRAPHITE 3726, GRAPHITE 3739,
7 GRAPHITE 5526, GRAPHITE 5539, GRAPHITE 9026, GRAPHITE 9039, and
8 GRAPHITE GA-17.

1 109. A removal agent for removing drag reducer from a liquid hydrocarbon

2 fuel, the removal agent comprising graphite selected from the group consisting of
3 GRAPHITE 2126, GRAPHITE 2139, GRAPHITE 3726, GRAPHITE 3739,
4 GRAPHITE 5539, GRAPHITE 9039, and GRAPHITE GA-17.

1 110. A removal agent for removing drag reducer from a liquid hydrocarbon
2 fuel, the removal agent comprising graphite selected from the group consisting of
3 GRAPHITE 2139, GRAPHITE 3726, GRAPHITE 3739, GRAPHITE 5539,
4 GRAPHITE 9039, and GRAPHITE GA-17.

1 111. A removal agent for removing drag reducer from a liquid hydrocarbon
2 fuel, the removal agent comprising graphite selected from the group consisting of
3 GRAPHITE 2139 and GRAPHITE 3739.

1

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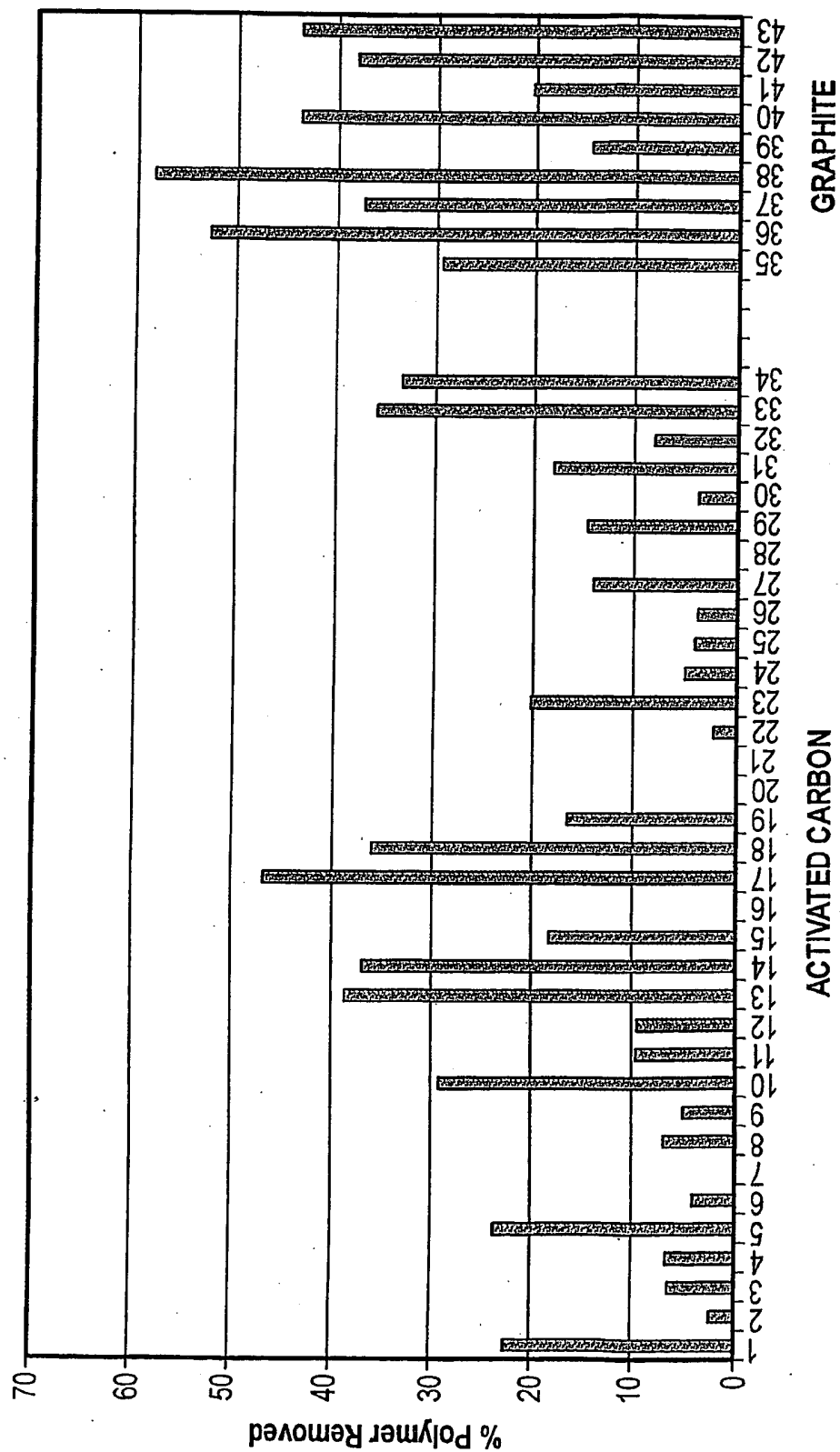


FIG. 1

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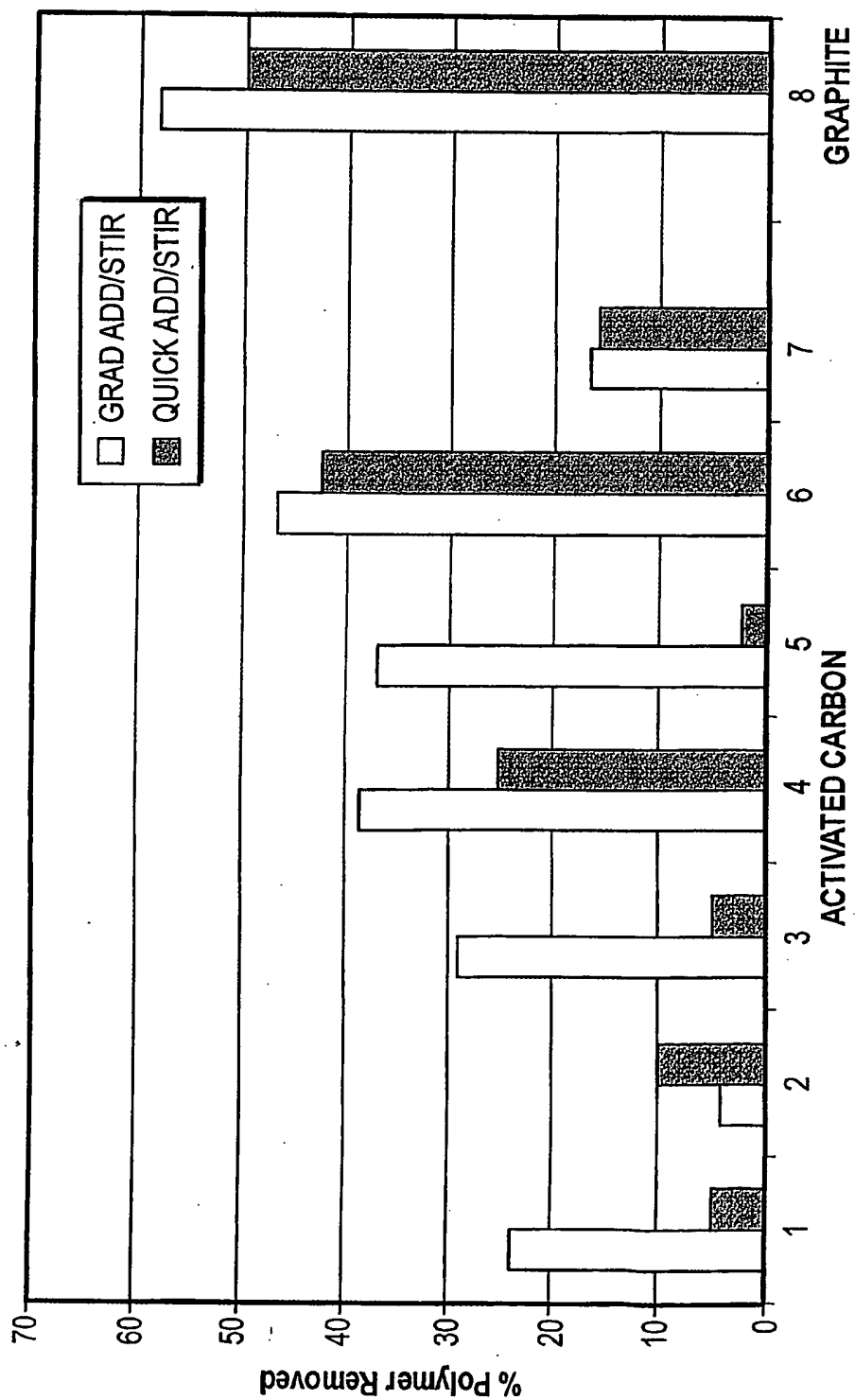


FIG. 2

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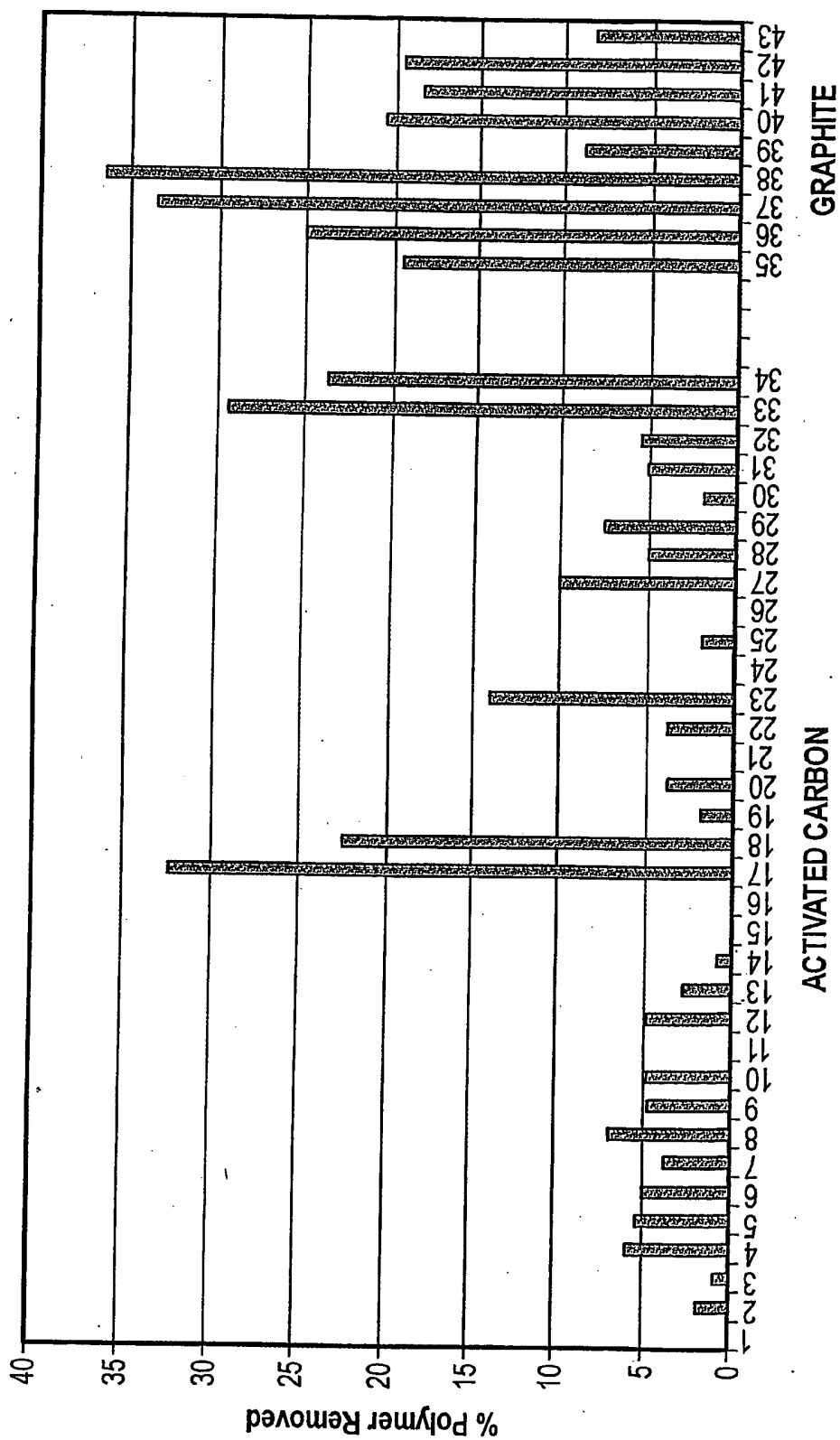


FIG. 3

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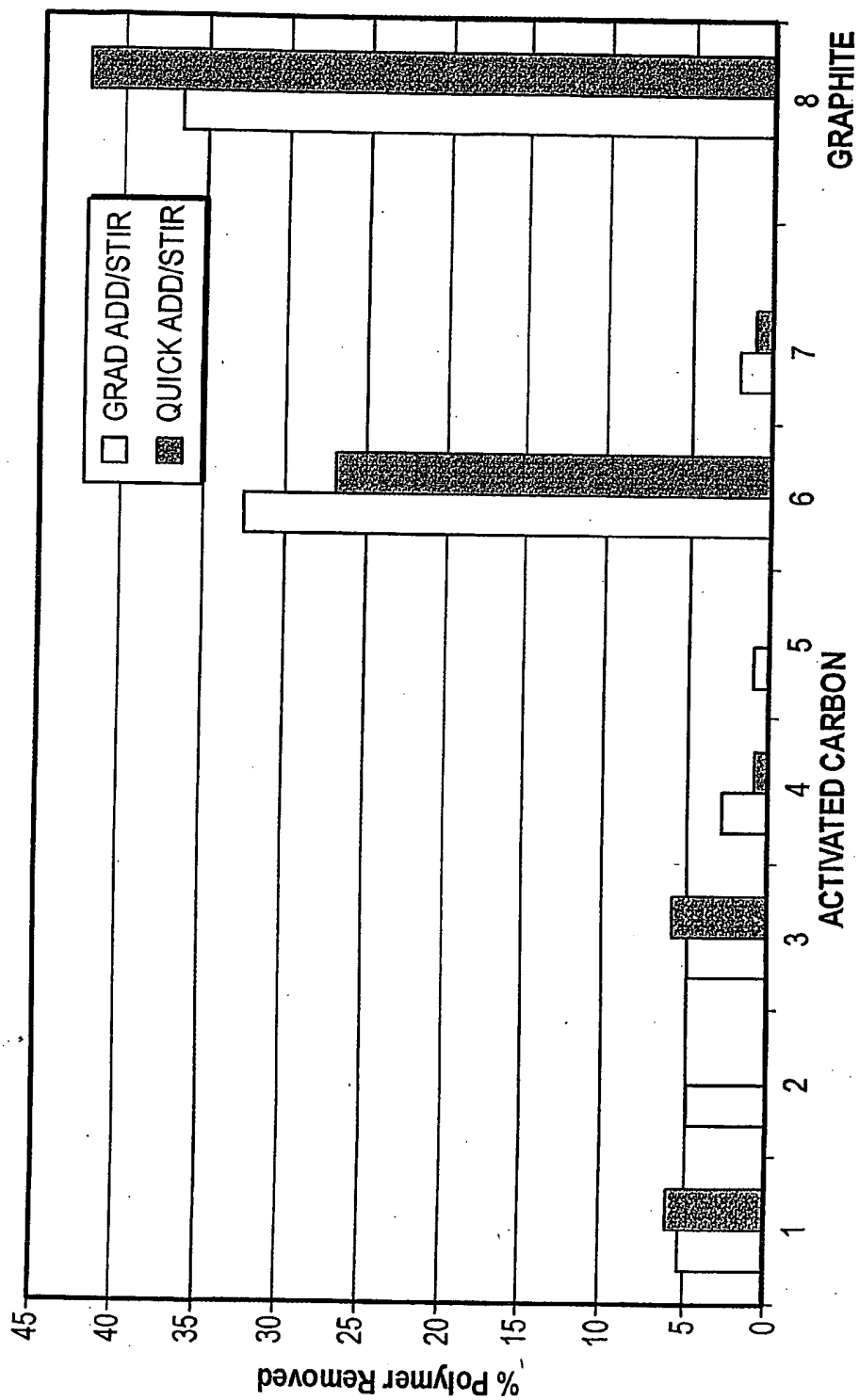


FIG. 4

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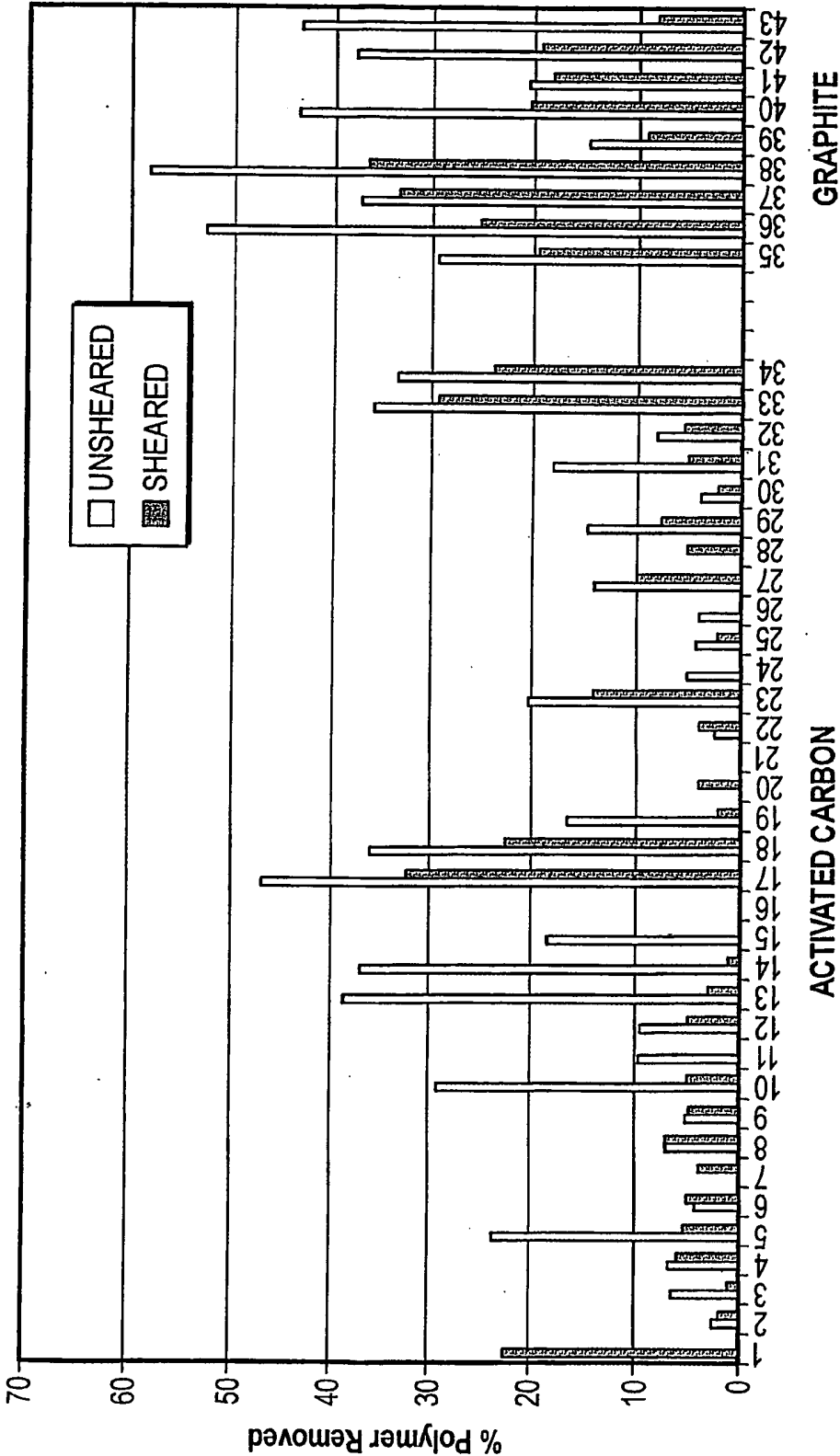


FIG. 5

INTERNATIONAL SEARCH REPORT

International Application No

PCT/US 03/17479

A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 C10G25/00 C10G25/06 C10G31/09

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 C10G C10L C01B B01J B01D

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal, WPI Data, PAJ

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	GB 1 236 066 A (BRITISH PETROLEUM CO LTD) 16 June 1971 (1971-06-16)	1,2
Y	claims 1-3,8,14 page 1, line 61-66 page 2, line 7-13 page 2, lines 96-103 and 114-120	1-16, 81-94
X	US 2003/019149 A1 (WAYNICK JOHN ANDREW) 30 January 2003 (2003-01-30) cited in the application	17-29, 81-104
Y	claim 1; figure 2; examples 1,2; table 1 page 1, paragraphs 12 and 16 page 2, paragraphs 18 and 21 page 3, paragraph 32 page 4, paragraph 42 page 5, paragraph 52	1-16, 81-94
	-/-	

☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

* Special categories of cited documents:

A document defining the general state of the art which is not considered to be of particular relevance

E earlier document but published on or after the international filing date

L document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

O document referring to an oral disclosure, use, exhibition or other means

P document published prior to the international filing date but later than the priority date claimed

T later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

X document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

Y document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

G document member of the same patent family

Date of the actual completion of the international search

12 February 2004

Date of mailing of the international search report

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INTERNATIONAL SEARCH REPORT

International Application No

PCT/US 03/17479

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US 4 837 249 A (O'MARA DION P ET AL) 6 June 1989 (1989-06-06) example 7 column 3, line 22-27 column 8, line 18-27	1-16, 81-94
A	US 5 788 865 A (GOLIPAD PYOTR NIKOLAEVICH ET AL) 4 August 1998 (1998-08-04) claim 1 column 5, line 12-19 column 6, lines 4-6, 25-27 and 45-48	1-16, 81-94
A	US 4 747 855 A (HIRAI HIDEFUMI ET AL) 31 May 1988 (1988-05-31) claims 1,2,5	1-16, 81-94

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US 03/17479

Box I Observations where certain claims were found unsearchable (Continuation of item 1 of first sheet)

This International Search Report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:
because they relate to subject matter not required to be searched by this Authority, namely:
2. ☐ Claims Nos.:
because they relate to parts of the International Application that do not comply with the prescribed requirements to such an extent that no meaningful International Search can be carried out, specifically:
3. ☐ Claims Nos.:
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

Box II Observations where unity of invention is lacking (Continuation of item 2 of first sheet)

This International Searching Authority found multiple inventions in this International application, as follows:

see additional sheet

As a result of the prior review under R. 40.2(e) PCT,
no additional fees are to be refunded.

1. ☐ As all required additional search fees were timely paid by the applicant, this International Search Report covers all searchable claims.
2. ☐ As all searchable claims could be searched without effort justifying an additional fee, this Authority did not invite payment of any additional fee.
3. ☒ As only some of the required additional search fees were timely paid by the applicant, this International Search Report covers only those claims for which fees were paid, specifically claims Nos.:

1-29, 81-104
4. ☐ No required additional search fees were timely paid by the applicant. Consequently, this International Search Report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

Remark on Protest

- ☒ The additional search fees were accompanied by the applicant's protest.
- ☐ No protest accompanied the payment of additional search fees.

FURTHER INFORMATION CONTINUED FROM PCT/ISA/ 210

This International Searching Authority found multiple (groups of) inventions in this international application, as follows:

1. Claims: 1-16,81-94

method of removing drag reducer additive (DRA) from a liquid hydrocarbon fuel comprising contacting said contaminated fuel comprising a concentration of a drag reducer additive with a quantity of one or more graphite effective to reduce said concentration of DRA

2. Claims: 17-29,81-94

method of removing drag reducer additive (DRA) from a liquid hydrocarbon fuel comprising contacting said contaminated fuel comprising a concentration of a drag reducer additive with a quantity of one or more activated carbon effective to having an adsorption capacity for DRA of 0.014 or more at from 8 to 12 ppm DRA and at 1g activated carbon/100 ml fuel, said DRA being sheared or unsheared

3. Claims: 30-43,81-94

method of removing drag reducer additive (DRA) from a liquid hydrocarbon fuel comprising contacting said contaminated fuel comprising a concentration of a drag reducer additive, said DRA comprising polyalphaolefin produced by solution polymerization, with a quantity of a removal agent selected from the group consisting of one or more graphite and one or more activated carbon having an adsorption capacity for said DRA of 0.014 or more at from 8 to 12 ppm DRA and at 1g activated carbon/100 ml fuel to produce a clean fuel

4. Claims: 44-80,81-94

method of removing drag reducer additive (DRA) from a liquid hydrocarbon fuel comprising contacting said contaminated fuel comprising a concentration of a drag reducer additive, said DRA comprising polymerized linear alpha olefin monomers having from 2 to 40 carbon atoms, with a quantity of a removal agent selected from the group consisting of one or more graphite and one or more activated carbon having an adsorption capacity for said DRA of 0.014 or more at from 8 to 12 ppm DRA and at 1g activated carbon/100 ml fuel to produce a clean fuel

5. Claims: 95-104

method for selecting a drag reducer additive (DRA) for a liquid hydrocarbon fuel comprising determining whether a single condidate DRA is removed from said fuel by a removal

FURTHER INFORMATION CONTINUED FROM PCT/SA/ 210

agent selected from the group consisting of graphite and activated carbon having an adsorption capacity for said DRA of 0.014 or more at from 8 to 12 ppm DRA and at 1g activated carbon/100 ml fuel and selecting as said DRA only candidates removed from said fuel by said removal agent

6. Claims: 105-107

removal agent comprising activated carbon selected from a group of commercial activated carbons

7. Claims: 108-111

removal agent comprising graphite selected from a group of commercial graphites

INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/US 03/17479

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US 4747855	A	31-05-1988	JP 1512896 C JP 60147239 A JP 63061053 B JP 1434527 C JP 60025939 A JP 62040333 B CA 1242684 A1 DE 3469411 D1 EP 0132915 A1	24-08-1989 03-08-1985 28-11-1988 07-04-1988 08-02-1985 27-08-1987 04-10-1988 31-03-1988 13-02-1985